Temperature Dependence of the Dielectric, Elastic and Piezoelectric Material Constants of Lead Zirconate Titanate Ceramics

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ABSTRACT

Smart structures based on piezoelectric materials are now finding applications in a wide variety of environmental conditions. We have used the impedance resonance technique to measure the impedance spectra of lead zirconate titanate ceramic specimens over temperatures ranging between 0ºC and 100ºC. The analysis of the radial, thickness, thickness shear and length extensional modes of resonance have allowed us to determine the temperature dependence of the complete reduced matrix of material constants, [s^E, ε^T, d], and our results are presented. We have also studied the effect of multiple heating cycles on the material and have observed the occurrence of thermal hysteresis. The causes and the significance of this hysteresis are analysed.

INTRODUCTION

Piezoelectric transducer materials are commonly characterised at room temperature by resonance measurements as outlined in the IEEE Standard on Piezoelectricity Std 176-1987 [1]. A small AC signal is used to excite a strain wave in the material via coupling to the piezoelectric coefficient. At a critical frequency determined by the sample dimensions a resonance occurs. The resonances are determined experimentally by monitoring the electrical impedance or admittance of the sample as a function of the frequency. The measurement is very practical in that the elastic, dielectric, and piezoelectric constants can be determined from one measurement [2]. However, in many applications, a transducer may be operated at temperatures above and below room temperature, depending on the application. It is therefore useful to know the temperature dependence of the material properties of the transducer material in order to predict the transducer response as a function of temperature. This paper reports on an investigation of the temperature dependence of complete reduced [s^E, ε^T, d] matrix for a PZT 4D type lead zirconate titanate ceramic and it includes a brief discussion about thermal hysteresis of the piezoelectric, dielectric, and elastic constants.

EXPERIMENT

Lead zirconate titanate ceramic (PZT 4D) manufactured by Morgan Matroc Ltd. was used to prepare specimens corresponding to the geometries outlined in the IEEE Standard on Piezoelectricity [1] to satisfy the boundary conditions required to measure the various modes of resonance. The resonance measurements were made using a HP 4194a impedance analyser with the necessary open/short circuit corrections carried out to eliminate parasitic impedances such as the parallel capacitance and the series resistance of the sample holder over the measurement frequency. The sample holder is placed in a Thermotron® programmable temperature chamber which allows the sample temperature to be adjusted between –72°C and +174°C. Impedance spectra of the radial, thickness, thickness shear, and length extensional modes of resonance were analysed using the impedance equations originally derived for free standing piezoelectric resonators by Berlincourt, Curran and Jaffe [3] and techniques reported earlier [1, 2, 4], and we have determined the 10 independent complex constants ($\mathbf{s}^E_{11}$, $\mathbf{s}^E_{12}$, $\mathbf{s}^E_{13}$, $\mathbf{s}^E_{33}$, $\mathbf{s}^E_{55}$, ε^T_{33}, ε^T_{11}, $\mathbf{d}_{33}$, $\mathbf{d}_{13}$, and $\mathbf{d}_{15}$) that define the reduced matrix for a piezoelectric material with C_∞ symmetry. The experiments were carried out at various temperatures between 0 C and 100 C and the variation of the material constants as a function of temperature was determined.

RESULTS AND DISCUSSION

Radial extensional mode (RAD)

A disc sample of diameter 30mm and thickness 0.7mm, which had an aspect ratio of radius to thickness greater than 15:1, was used to study the radial mode of resonance. The impedance spectra were analysed using the technique of Sherrit et al. [2]. The temperature dependence of the elastic, dielectric and piezoelectric constants ($\mathbf{s}_{11}^E$, $\mathbf{s}_{12}^E$, ε_{33}^T, $\mathbf{d}_{13}$) governing the radial mode are shown in Figure 1.

Thickness extensional mode (TE)

The same disc sample was used to determine the thickness mode resonance. The results were analysed to find the value of the open circuit elastic stiffness $\mathbf{c}_{33}^D$.

Length extensional mode (LE)

The sample used was a rod of length 10.8mm and width of 4.2mm and 1.6mm. The temperature dependence of the material constants ($\mathbf{s}_{33}^E, \mathbf{k}_{33}$) governing the length extensional mode are plotted in Figure 1. The IEEE standard for the length extensional mode recommends that the sample's length be at least five times its lateral dimension. The aspect ratio for our sample was of the order of 3:1, which is lower than the recommended value and so the values quoted for $\mathbf{d}_{33}$ may be slightly lower than the true values, due to lateral clamping. The value of ε_{33}^T determined from this mode is not reported since this value can be determined more accurately from an analysis of the radial mode. The value of ε_{33}^T determined using the length extensional resonator was however within 10% of the values determined by the radial and the length thickness modes.

Thickness shear extensional mode (TS)

The sample used was a plate 10.6mm x 3.3mm x 1.02mm. The temperature dependence of the material constants ($\mathbf{s}_{55}^E, \varepsilon_{11}^T, \mathbf{k}_{15}$) governing the thickness shear mode are plotted in Figure 1.

Determination of $\mathbf{s}_{13}^E$

The value of $\mathbf{s}_{13}^E$ was determined by inverting the 9x9 [s^E, ε^T, d] matrix with an initial guess for the value of $\mathbf{s}_{13}^E$ to get the [c^D, β^S, h] matrix. The value of $\mathbf{s}_{13}^E$ was adjusted until the value of $\mathbf{c}_{33}^D$ determined from the inverted matrix equalled the value of $\mathbf{c}_{33}^D$ determined from the thickness resonance analysis. The calculations used the material constants determined from the fundamental resonances of all the modes discussed above. The temperature dependence of the value of $\mathbf{s}_{13}^E$ is also shown in Figure 1. $\mathbf{s}_{13}^E$ can also be calculated using the equations described by Smits [4] and there is good agreement between the values obtained by using the two different methods.

Polynomial fits

In order to help transducer designers, we have expressed the experimental values of the material constants as polynomials in temperature and these are listed in Table 1. These polynomials provide good fits to the experimental data.

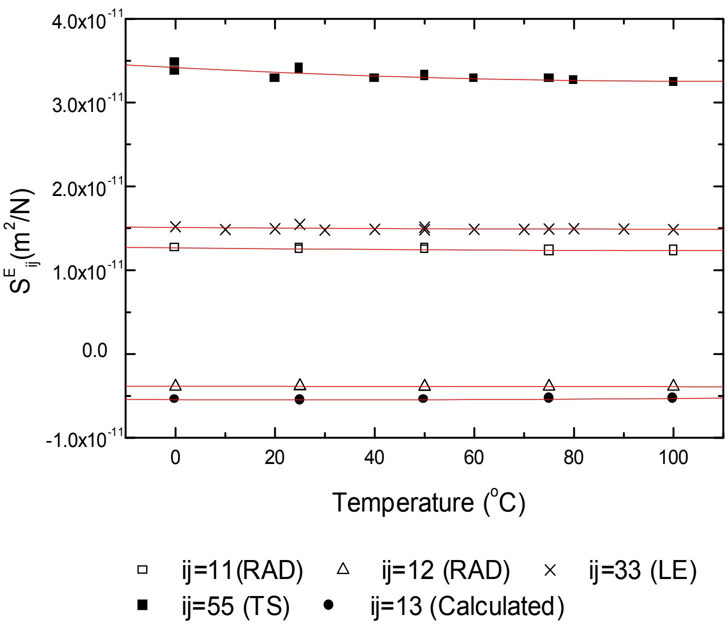


Figure 1(a): Temperature dependence of the elastic constants.

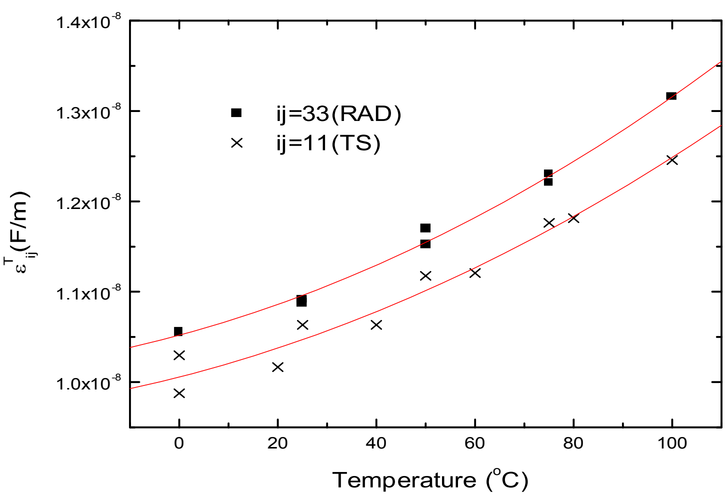


Figure 1(b): Temperature dependence of the dielectric coefficients.

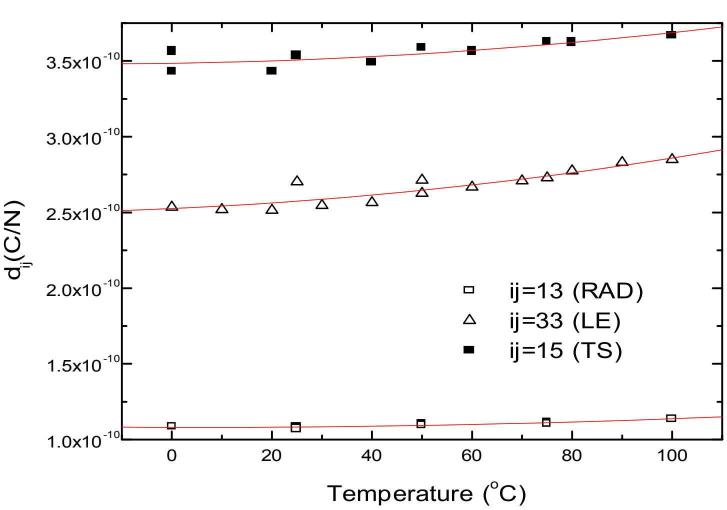


Figure 1(c): Temperature dependence of the piezoelectric d coefficients.

Table 1: The polynomial fit to the data shown in Figure 1

Material Constant (Resonance used)	Polynomial (T in °C)
S^E_{11}(RAD) (m^2/N)	$=1.27x10^{-11}-5.39x10^{-15}T+2.23x10^{-17}T^2$
S^E_{12}(RAD) (m^2/N)	$=-3.86x10^{-12}-2.17x10^{-16}T-2.06x10^{-18}T^2$
S^E_{33}(LE) (m^2/N)	$=1.51x10^{-11}-4.03x10^{-15}T+1.94x10^{-17}T^2$
S^E_{55}(TS) (m^2/N)	$=3.42x10^{-11}-3.07x10^{-14}T+1.43x10^{-16}T^2$
S^E_{13}(Cal) (m^2/N)	$=-5.44x10^{-12}-1.92x10^{-15}T+3.20x10^{-17}T^2$
ε^T_{33}(RAD) (F/m)	$=1.05x10^{-8}+1.47x10^{-11}T+1.17x10^{-13}T^2$
ε^T_{11}(TS) (F/m)	$=1.01x10^{-8}+1.39x10^{-11}T+1.04x10^{-13}T^2$
d_{13}(RAD) (C/N)	$=1.08x10^{-10}-7.09x10^{-15}T+6.38x10^{-16}T^2$
d_{33}(LE) (C/N)	$=2.53x10^{-10}+1.49x10^{-13}T+1.85x10^{-15}T^2$
d_{15}(TS) (C/N)	$=3.49x10^{-10}+4.90x10^{-14}T+1.53x10^{-18}T^2$

The mechanical Q of the material

The variation of the mechanical Q as a function of temperature has been found to be highly dependent on the mode of excitation as shown in figure 2. The mechanical Q of the length extensional mode is seen to be 100 and decreases substantially as the temperature is increased. The mechanical Q of the elastic constants governing the radial mode and the thickness mode are of the order of 200 and are seen to increase as the temperature increases. This suggests that the proper damping coefficients should be carefully chosen for practical applications.

Dielectric dissipation

Figure 3 shows the electrical dissipation for the thickness shear and radial mode resonator at and below resonance. It should be noted that, like the mechanical Q, the electrical loss is dependent on the direction of the excitation. The electric dissipation determined at resonance is seen to be measurably lower than the data determined at 10 kHz, which indicates significant frequency dispersion in the loss.

Thermal hysteresis

The effect of multiple heating cycles on the material constants has been investigated. Two sets of radial mode data ($\mathbf{s}^E_{11}$, $\mathbf{s}^E_{12}$, ε^T_{33}, $\mathbf{d}_{13}$) under two consecutive heating cycles are shown in Figure 4. A significant difference was found between the first cycle and later cycles. A hysteresis exists in the material constant versus temperature curves. In typical ferroelectric materials a measurable thermal hysteresis can be expected due to irreversible domain wall motions in the material [5]. The 180° and 90° domain walls move to so as to minimise the domain energy and some of the domains engulf other domains or change shape irreversibly thus contributing to the net strain and polarisation and resulting in a measurable hysteresis. The hystereses in later cycles are significantly reduced since the contribution due to the irreversible motion in is much less in later cycles than in the first cycle. For applications at elevated temperature we recommend that the material be heat treated prior to characterisation so as to reduce the contribution of the irreversible processes and the thermal hysteresis.

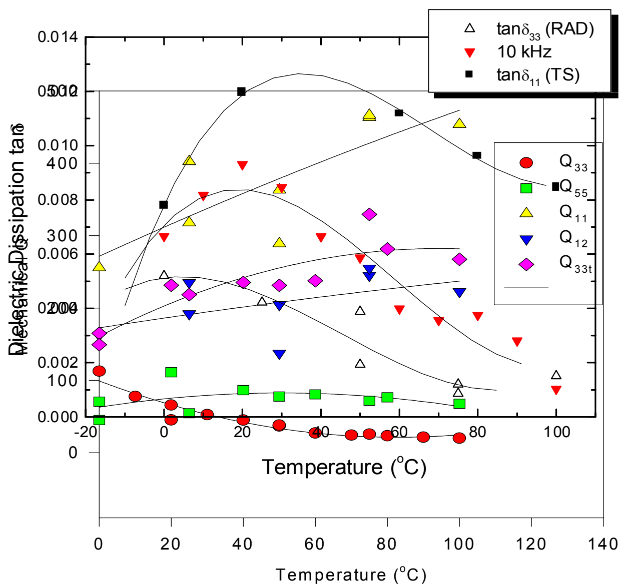
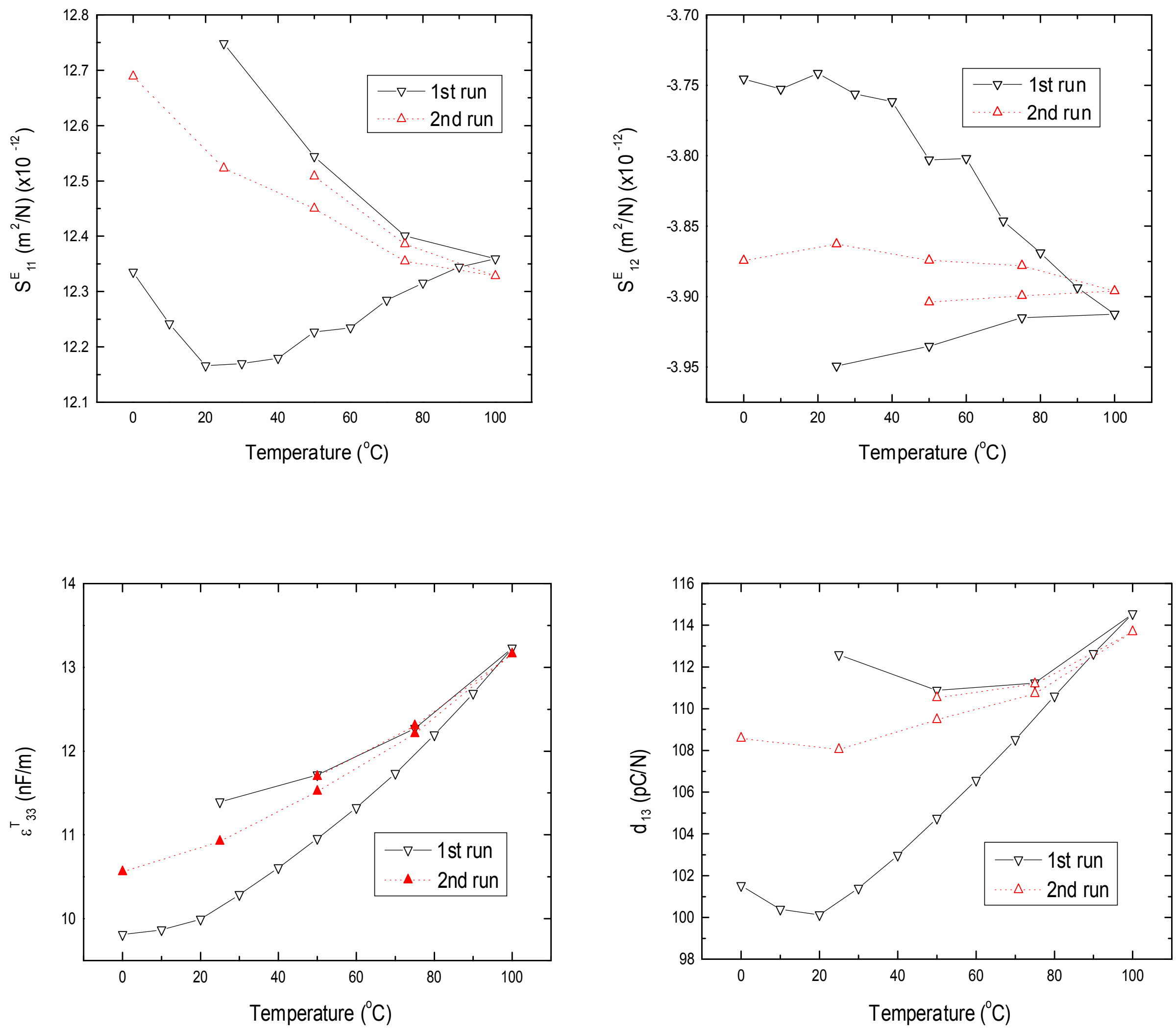


Figure 2: Temperature dependence of the mechanical Q of the material.

Figure 3: Temperature dependence of the dielectric dissipation tanδ.

CONCLUSION

Four common resonance modes for PZT-4D piezoelectric ceramic produced by Morgan Matroc Ltd. were measured and analysed in the temperature range from 0°C to 100°C. The 10 independent material constants ($\mathbf{s}_{11}^{E}$, $\mathbf{s}_{12}^{E}$, $\mathbf{s}_{13}^{\mathrm{E}}$, $\mathbf{s}_{33}^{E}$, $\mathbf{s}_{55}^{E}$, ε_{33}^{T}, ε_{11}^{T}, $\mathbf{d}_{33}$, $\mathbf{d}_{13}$, and $\mathbf{d}_{15}$) that define the reduced matrix for a C_{∞} piezoelectric material were determined from analysis of the fundamental resonance in various modes. Polynomials were used to fit the data so that the data could be reported in tabular form. The polynomials were found to represent the data well over the entire temperature range of measurement. A significant thermal hysteresis in the material constants as a function of temperature was observed. The effect of the heating process on the resonators has to be taken into account if they are to be used at elevated temperature. The results suggest that heat treating the material reduces the subsequent hysteresis.



ACKNOWLEDGEMENT

The authors thankfully acknowledge the financial support provided by Chief of Research and Development, the Department of National Defence, Canada and by the US Office of Naval Research through the Naval Underwater Warfare Center, Newport, RI, USA.

Figure 4: Thermal behaviour of the material constants under two consecutive heating cycles.

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